U.S. DEPARTMENT OF COMMERCE
National Technical Information Service

PB-268 960

Evaluation of the Technological Feasibility, and Cost of Selected Control Alternatives Necessary to Meet the Proposed Ohio SO2 Regulations for Industrial Boilers and Processes. Volume VII. The Standard Oil Company of Ohio, Lima Refinery

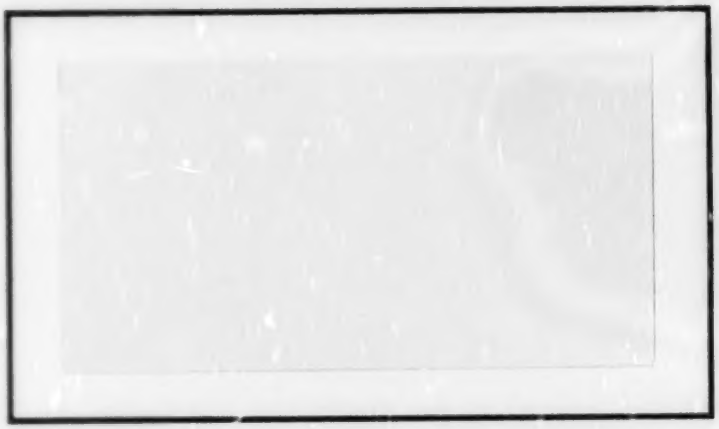
GCA Corp, Bedford, Mass. GCA Technology Div.

Prepared for

Environmental Protection Agency, Chicago, Ill. Air and Hazardous Materials Div.

Aug 76

PB 268 960



GCA/TECHNOLOGY DIVISION

EPA-905/2-76-005-g

GCA-TR-76-14-G(7)

EVALUATION OF THE TECHNOLOGICAL FEASIBILITY, AND COST OF SELECTED CONTROL ALTERNATIVES NECESSARY TO MEET THE PROPOSED OHIO SO_2 REGULATIONS FOR INDUSTRIAL BOILERS AND PROCESSES

FINAL REPORT

Contract No. 68-01-3155
Task Order No. 4

August 1976

Volume VII

THE STANDARD OIL COMPANY OF OHIO,
LIMA REFINERY

by

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Washington, D.C. 20460

TECHNICAL REPORT DATA (Please read Instructions on the reverse before completing)		
1. REPORT NO. EPA-905/2-76-005-g	2.	3. RECIPIENT'S ACCESSION NO.
4. TITLE AND SUBTITLE Evaluation of the Technological Feasibility and Cost of Selected Control Alternatives Necessary to Meet the Proposed Ohio SO_2 Regulations for Industrial Boilers and Processes. Volume VII		5. REPORT DATE August 1976 6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) Walter T. Stanley Robert R. Hall		8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS GCA Corporation GCA/Technology Division Bedford, Massachusetts		10. PROGRAM ELEMENT NO. 11. CONTRACT/GRANT NO. 68-01-3155 Task Order No. 4
12. SPONSORING AGENCY NAME AND ADDRESS Air Programs Branch, A&HMD Environmental Protection Agency, Region V 230 S. Dearborn Chicago, Illinois 60604		13. TYPE OF REPORT AND PERIOD COVERED Final 14. SPONSORING AGENCY CODE
15. SUPPLEMENTARY NOTES One of 9 documents supporting sulfur dioxide control strategy for the State of Ohio Technical Support Document Volumes I and II EPA 905/2-76-002		

16. ABSTRACT

This report presents an evaluation by the GCA Corporation, GCA/Technology Division, of technological feasibility and cost of complying with the proposed Ohio sulfur dioxide regulation (40 FR 52410, November 10, 1975) at the Standard Oil Company of Ohio's Lima Refinery. The final USEPA Sulfur Dioxide Control Strategy for the State of Ohio Technical Support Documents, Vol. I and II, (EPA 905/2-76-002) may be obtained from NTIS.

After collecting necessary site and process specific data by means of a plant visit, it was determined that the only source not meeting the proposed regulation was the Claus sulfur recovery plant. For this plant, the technical feasibility and cost of compliance using the Beavon, IFP-150, IFP-1500, Shell Claus off-gas treating (SCOT), sulfreen, and Wellman-Lord processes for desulfurization of the tail gases were evaluated.

17. KEY WORDS AND DOCUMENT ANALYSIS		
a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Sulfur Sulfur Dioxide	Control Strategy Sulfur Recovery Plants Ohio Standard Oil Company of Ohio	13B 07B
18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES 51
	20. SECURITY CLASS (This page)	22. PRICE A04-A01

EPA Form 2220-1 (9-73)

This Final Report was furnished to the Environmental Protection Agency by the GCA Corporation, GCA/Technology Division, Bedford, Massachusetts 01730, in fulfillment of Contract No. 68-01-3155, Task Order No. 4. The opinions, findings, and conclusions expressed are those of the authors and not necessarily those of the Environmental Protection Agency or of the cooperating agencies. Mention of company or product names is not to be considered as an endorsement by the Environmental Protection Agency.

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ACKNOWLEDGMENTS

The assistance of Mr. Wayne E. Pearson, Project Officer, and Mr. Thomas Blaszak, U.S. Environmental Protection Agnecy, Air Policy Branch, Region V, Chicago, Illinois is gratefully acknowledged.

SECTION I

SUMMARY AND CONCLUSIONS

The Standard Oil Company of Ohio (SOHIO) operates an oil refinery in Lima, Ohio, with a processing capacity of 180,000 barrels of crude oil per day. The current emission levels from numerous sources were determined and checked for compliance with the proposed Ohio SO_2 emission regulations. The only source not meeting the proposed limitations was the Claus sulfur recovery plant which would be limited to 0.01 lbs SO_2/lb sulfur processed. The technical feasibility, and cost, of reducing SO_2 emissions to this level (a 91 percent reduction) were evaluated.

The following commercially available Claus plant tail-gas desulfurization processes were investigated:

- Beavon sulfur removal process
- IFP - 150 process
- IFP - 1500 process
- Shell Claus off-gas treating (SCOT) process
- Sulfreen process
- Wellman-Lord SO_2 recovery process

A discussion of each process appears in the body of this report. All systems except the IFP - 1500 process and Sulfreen process are technically capable of meeting the proposed standard. The total capital requirement and annual operating costs for a retrofit Beavon system and SCOT system were evaluated. The required capital investment is $2.0 million for a Beavon unit and $2.1 million for a SCOT unit. The associated annual costs are $541,000 and $652,000, respectively. These

annual costs are equivalent to $0.346 and $0.429 per pound of sulfur removed.

The 1976 capital requirement and annual operating costs for a fuel gas desulfurization and sulfur recovery facility equivalent to that in use at the Lima refinery were also estimated. These costs are of considerable interest to the oil companies in Ohio who are operating refineries burning refinery fuel gas which contains more than the 50 grains H_2S/100 scf limit proscribed in the proposed regulations. The total capital investment for such a system would be $8.6 million with an annual cost of $3.3 million which is equivalent to $0.12 per pound of sulfur removed. The above system does not include Claus tail-gas treatment which would require an additional capital investment of about $2 million and annual costs of $600,000 or less

SECTION II

PROCESS AND SITE DESCRIPTION

The Lima Refinery of The Standard Oil Company of Ohio (SOHIO) was visited on 27 April 1976. This facility is located on the outskirts of Lima, Allen County, Ohio, on the Ottawa River. With a processing capacity of 180,000 barrels of crude oil per day, it is the largest refinery in the state. The purpose of the plant visit was to collect relevant operating data to determine the technical feasibility and cost of complying with the proposed Ohio SO_2 emission regulations. Mr. P. C. Tranquill and Mr. W. H. Connell represented SOHIO. The refining facilities and operation were discussed, and process data, fuel analyses, flow diagrams, and site layout drawings were provided by SOHIO. Ford, Bacon, and Davis, Inc. of Dallas, Texas, was contacted during the interview for release of information concerning the modified Claus sulfur recovery unit which they designed and built for SOHIO.

The Lima Refinery utilizes a variety of unit operations to process crude oil into fuels, lubricants, and other petroleum products. Due to its complexity, the refinery has many SO_2 emission points.

The principal sources are the following:[1]

1. Refinery process heaters and boilers
2. Sulfur recovery plant
3. Catalytic cracker/CO boiler off-gas

EMISSIONS FROM COMBUSTION SOURCES

The process heaters and boilers burn a variety of fuels. These include natural gas, liquid petroleum gas (LPG), refinery fuel gas, and No. 6 residual fuel oil. The natural gas is supplied by an outside source, and the remaining fuels are produced at the refinery. The natural gas is essentially sulfur-free. The refinery fuel gas, LPG, and residual oil contain sulfur which originates from the crude oil. Sulfur content of the crude oil averages 0.4 percent by weight, with a maximum of 0.5 percent.[2]

The proposed sulfur dioxide emission regulations limit emissions from refinery combustion sources by three methods.[3] Section 52.1881(b)(9) restricts the sulfur content of refinery fuel gas as follows:

> (9) Except as provided in paragraph (b)(7) of this section no owner or operator of a petroleum refinery shall cause or permit the combustion of fossil fuel containing a total sulfur content expressed as hydrogen sulfide (H_2S) in excess of 50 grains per dry standard 100 cubic feet of fossil fuel combusted.

The Standard Oil Company (SOHIO) assumes that this limit applies only to gaseous fuels generated by refinery process units.[4] GCA concurs with this assumption. Boilers burning liquid or solid fossil fuel would be subject to an emission limitation of 1.6 lb $SO_2/10^6$ Btu of actual heat input. This is the county-specific boiler emission limit presented in §52.1881(b)(2). It also appears that emissions from process heaters firing liquid or solid fossil fuels will be limited by the process weight equation which appears in §52.1881(b)(8).

> (8) Except as provided in paragraphs (b)(7),(9),(10),(11), (12), and (13) of this section, no owner or operator of any process equipment shall cause or permit the emission into the atmosphere of any process gas stream containing sulfur dioxide in excess of the amount prescribed by the following equation:
>
> $$E = 19.5p^{0.67}$$
>
> where E is the allowable emission rate in lbs SO_2/hour and p is the tons of process weight per hour.

Coal is not presently being burned at the Lima Refinery. For process heaters firing No. 6 residual oil, the process weight equation leads to emission limitations ranging from 2.6 to 21.5 lb $SO_2/10^6$ Btu.[4] According to fuel oil analyses furnished by SOHIO in their response to the Section 114 information request, the Lima Refinery burns oil with a mean sulfur content of 0.84 weight percent. The heating value of the fuel is 144,000 Btu/gal. Combustion of this oil results in sulfur dioxide emissions of 0.90 lb $SO_2/10^6$ Btu, well in compliance with both the process regulation and the boiler regulation. These figures are substantiated by the Ohio EPA Emissions Inventory Confirmation Report.

The refinery gas and LPG are treated in the refinery gas desulfurization and sulfur recovery system before they are burned. A schematic of the process is provided in Figure 1. Two basic operations are involved in this treatment system. First the scur gases are contacted with an amine solution which absorbs the H_2S. The sulfide-rich amine solution is then stripped with steam and the resulting acid gas is fed into a Claus sulfur recovery unit. In addition, acid gas from a foul condensate stripping unit is fed to the Claus plant.

There are two amine desulfurization sections in the process. One unit desulfurizes sour fuel gas using an 18 percent diethanolamine solution (DEA) as the absorption medium. The DEA treated fuel gas originates from the Fluid Catalytic Cracking Unit, the Delayed Coking Unit, and the Thermal Gas Plant. Sour propylene from the catalytic cracker is also desulfurized in a DEA section. Sulfide-rich DEA from both the propylene contactor and the refinery fuel gas contactor is regenerated with steam and the lean amine solution is recycled to the contactors. The acid gas is sent to the Claus plant. Flow rates and sulfur content of the gas streams to the DEA section are shown in Table 1.[2,5] The heating value of the treated fuel gas is 1171 Btu/scf. Desulfurization of the DEA-treated gas was initiated in June 1974.[2]

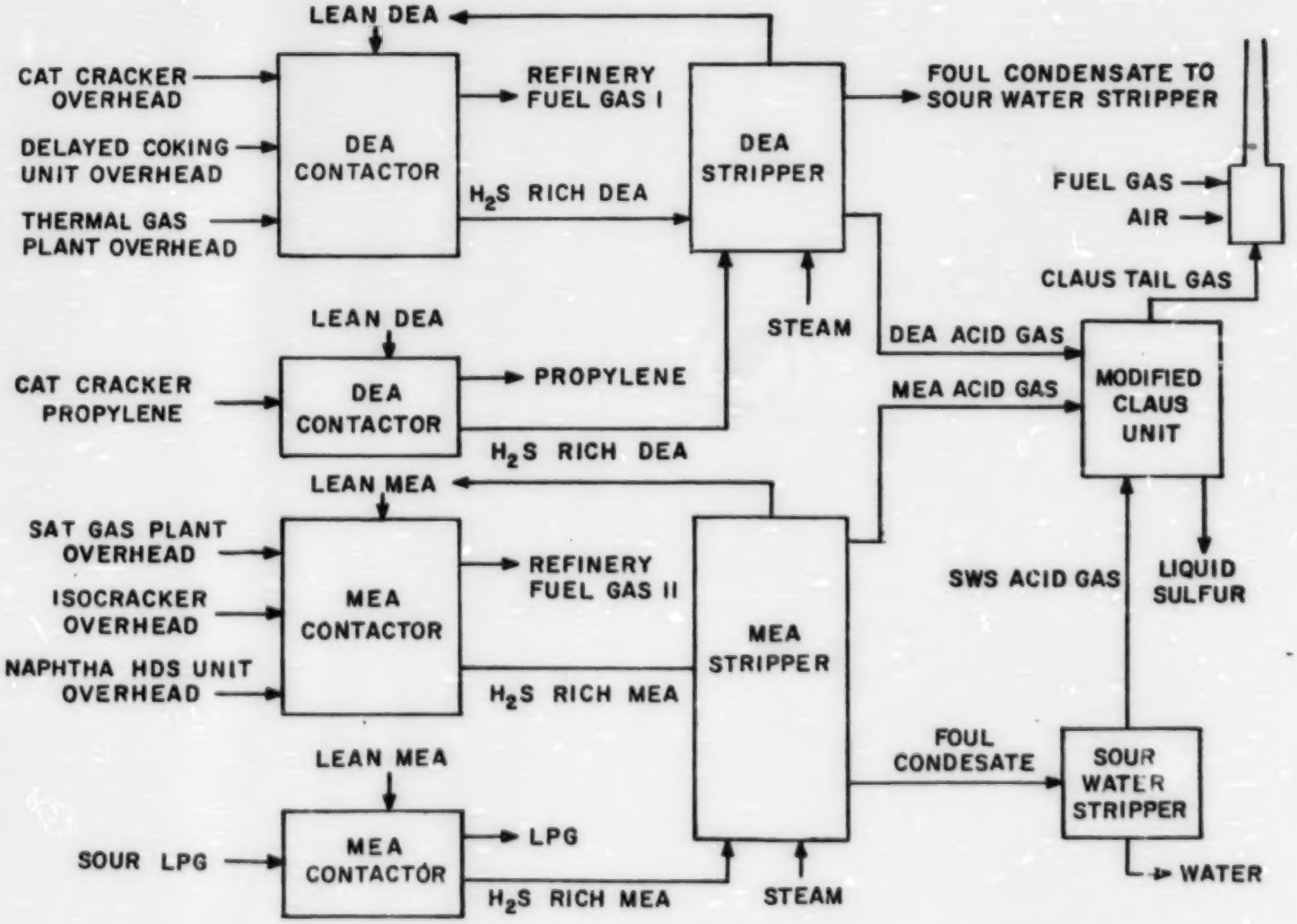

Figure 1. Refinery gas desulfurization and sulfur recovery system[2]

Table 1. FLOW RATES AND SULFUR CONTENT OF STREAMS TO AMINE CONTACTORS[2,5]

	Flow rate, 10^3 scf/hr		Sulfur content before treatment, gr H_2S/100 scf
	Norm	Design max	
DEA unit			
Refinery fuel gas I	488	600	1,700
Propylene	178	233	1,400
MEA unit			
Refinery fuel gas II	350	475	1,420
LPG	243	327	520

The second amine desulfurization section treats fuel gases from the Sat Gas Plant, Isocracker, and Naphtha HDS Unit. The gases are combined and fed to a monoethanolamine (MEA) contactor. LPG from the Sat Gas Plant is also scrubbed with MEA solution in a separate contactor. The rich MEA is then sent to an amine stripping unit where acid gas is removed from the MEA solution and sent to the sulfur recovery plant. The MEA solution is recycled to the contactors. Flow rates and sulfur content of streams entering the MEA contactors are shown in Table 1.[2,5] The MEA-treated fuel gas has an average heating value of 1097 Btu/scf.[2]

Based on analyses performed by Lima Refinery personnel, the amine treating units are currently desulfurizing the fuel gas to well within regulatory limits. These analyses were performed in conjunction with four sulfur recovery unit environmental compliance tests. Test I was done as a preliminary evaluation of the unit operations. Test II was conducted under the observation of an Ohio EPA representative. Results of the tests are summarized in Table 2.[5] The fuel gas was sampled and analyzed according to U.S. EPA Method 11 (modified), "Determination of Hydrogen Sulfide Emissions from Stationary Sources." Although less precise, the Tutweiler technique for determining the quantity of H_2S in fuel gas was used to confirm the findings obtained using the modified Method 11.

Table 2. SUMMARY OF FUEL GAS SULFUR ANALYSES[5]

Test date	Sulfur content of treated fuel gas grains H_2S equivalent/100 scf			
	Dec. 12-13, 1974	April 10, 1975	July 16, 1975	Sept. 24, 1975
DEA treated fuel gas (RFG I)	45	6	4.4[a]	8
MEA treated fuel gas (RFG II)	20	11	not detected[a]	24

[a] H_2S determination was made by Tutweiler technique only.

Treated refinery fuel gas always contained less than the 50 grain H_2S equivalent/100 scf. Hence, all combustion sources are in compliance with the proposed SO_2 emission regulations.[5]

SULFUR RECOVERY PLANT EMISSIONS

As shown in Figure 1, acid gas streams from the DEA unit, MEA unit, and the sour water stripper are sent to a Claus plant to recover sulfur. The plant is a modified 3-stage system engineered by Ford, Bacon, and Davis, Inc., and built in 1974. Production capacity is 33.6 long ton sulfur/day when operating at the guaranteed 94.5 percent recovery efficiency. SO_2 emissions would be 1600 short tons per year. The acid gas flow rates and compositions used for the design of the Claus unit are presented in Table 3.[2,6]

Currently, the sulfur recovery unit is producing approximately 30 long tons/day of sulfur. The sulfur is greater than 99 percent pure and is produced in a molten state. The sulfur is sold to a sulfuric acid manufacturing company for $45/lt.[2]

Sulfur dioxide emission in Claus plant tail-gas is limited by §52.1881(b)(13)[3] of the proposed regulations which states:

Table 3. FLOW RATE AND COMPOSITION OF ACID GAS STREAMS TO CLAUS PLANT, DESIGN BASIS[2,6]

Component	DEA unit acid gas stream	MEA unit acid gas stream	Sour water stripper acid gas stream	Total	
	Moles/hr	Moles/hr	Moles/hr	Moles/hr	Vol. %
Hydrogen sulfide, H_2S	55.23	35.32	11.76	102.31	51.13
Carbon dioxide, CO_2	35.10	0.90	0.40	36.40	18.19
Carbon monoxide, CO	0.27	0.01	-	0.28	0.14
Hydrogen, H_2	0.17	0.32	-	0.49	0.24
Nitrogen, N_2	0.14	-	-	0.14	0.07
Hydrocarbons	2.73	3.16	0.98	6.87	3.43
Ammonia, NH_3	0.15	-	23.53	23.68	11.83
Water, H_2O	9.13	3.88	15.70	28.71	14.35
Methyl mercaptan +, CH_3SH	1.04	-	-	1.04	0.52
Carbonyl sulfide, COS	0.21	-	-	0.21	0.10
Total	104.17	43.59	52.33	200.09	100
LT/D sulfur in feed gas	19.41	12.13	4.04	35.58	

> (13) No owner or operator of a sulfur recovery plant shall cause or permit the emission of sulfur dioxide from the plant in excess of 0.01 lb SO_2/lb of sulfur processed. The provisions of this subparagraph shall not be applicable to processes operated in conjunction with the desulfurization of byproduct coke oven gas provided the emission limitation set forth in paragraph (b) (12) of this section has been complied with.

However, it is not clear whether the above limitation applies to the Lima Refinery Claus plant. According to Paragraph XIV of Section 52.1881(b) (1), sulfur recovery plant is defined as follows:

(xiv) "Sulfur recovery plant" means any plant that recovers elemental sulfur from any gas stream, but not include facilities where sulfur recovery is utilized primarily as a means of preventing emissions to the atmosphere of sulfur dioxide or other sulfur compounds.

The SOHIO Claus plant is part of the refinery fuel gas desulfurization system, and as such might be excluded from compliance with the sulfur recovery plant emission standard. In the event the regulation does apply to refinery sulfur recovery units, a Claus plant tail-gas treatment unit would be needed. At present, the Claus plant is operating at approximately 96 percent sulfur recovery efficiency, emitting 0.08 lb SO_2/lb sulfur processed. These operating characteristics are substantiated by the results of the environmental compliance tests previously mentioned. The results of the tests are reported in Table 4.[5]

Table 4. SUMMARY OF CLAUS PLANT SULFUR OXIDE EMISSION TESTS[5]

	Test			
	Dec. 12-13, 1974	Apr. 10, 1975	July 16, 1975	Sept. 24, 1975
Sulfur oxide emission rate				
lb SO_2 equivalent/hr	214/186	142	254	359[a]
lb SO_2 equivalent/lb sulfur processed	0.072/0.063	0.051	0.088	0.12[a]
Sulfur production rate				
lb/hr	2860	2710	2757	2810
LT/day	30.6	29.0	29.5	30.1
Recovery efficiency, %	96.4	97.5	95.6	94.0[a]

[a]Sampling was done after interruption of unit operations before steady state conditions had been attained.

As previously mentioned, the guaranteed recovery efficiency of the modified Claus unit is 94.5 percent or 0.11 lb SO_2 emitted/lb sulfur processed. Hence, the tail-gas desulfurization unit must be designed to recover 91 percent of the sulfur currently being emitted to the atmosphere. This amounts to 163 lb sulfur/hr at the maximum production rate. These points are illustrated in Table 5.

Table 5. DESIGN PERFORMANCE OF CLAUS AND TAIL GAS TREATMENT UNITS

	Design-Claus unit	Allowable per §52.1881(b)(13)	Design-tail-gas unit	Design-combined Claus and tail-gas units
Maximum sulfur dioxide emission rate				
lb SO_2/hr	360	33.2	33	33
lb SO_2/lb sulfur processed	0.11	0.01	0.18	0.01
Sulfur production rate				
lb/hr	3137	-	163	3300
LT/day	33.6	-	1.8	35.4
Recovery efficiency (minimum), %	94.5	99.5	91	99.5

Requests for quotations on a turnkey installed battery-limit plant were submitted to vendors of Claus tail-gas desulfurization systems. The facility was to be designed to meet the following process specifications in addition to the proposed regulation.

1. Feed stream characteristics (Claus unit tail-gas)[2,6]
   - Volumetric flow rate, scf/hr 227,500
   - Mass flow rate, lb/hr 16,300
   - Temperature, ^{o}F 310
   - Pressure, psia 17.7

- Chemical composition

	lb-mole/hr	Vol. %
N_2	363.27	57.30
H_2	200.17	31.58
CO_2	38.63	6.09
CO	20.92	3.30
H_2	5.43	0.86
H_2S	2.10	0.33
S (vapor)	1.54	0.24
SO_2	1.05	0.17
CH_3SH	0.50	0.08
S (liquid)	0.26	0.04
CS_2	0.08	0.01
Total	663.95	100.

- Total sulfur, lb/hr 179.5

2. Off gas from the desulfurization unit shall contain no more than 0.18 lb SO_2/lb sulfur in feed (Claus unit tail-gas)
3. Recovered sulfur shall be in elemental molten form
4. Adequate space is available near Claus unit
5. Available utilities[2]
   - Refinery fuel gas, 1170 Btu/scf
   - Electric power, any voltage
   - Steam: 10 psig, 40 psig, 110 psig, saturated; 600 psig superheated
   - Water

A description and technical evaluation of available Claus tail-gas desulfurization systems appears in Section III. The estimated capital and annualized costs of such systems are presented in Section IV.

FLUID CATALYTIC CRACKER REGENERATOR/CO BOILER EMISSIONS

Fluid catalytic cracking (FCC) plants and the associated carbon monoxide boilers typically emit significant quantities of sulfur dioxide.[1] The term fluid refers to the silica-alumia catalyst which as a dust flows through the system like a liquid. Feedstocks to the FCC unit are heavy oils from the crude oil fractionation tower and delayed coker units.[2] The large hydrocarbon molecules are pyrolyzed into lighter components such as gasoline and fuel oil in the presence of the fluidized catalyst. The catalyst must be periodically regenerated to remove coke deposits. This is accomplished in the FCC regenerator where the coke is oxided with air and the bulk of the catalyst returned to the reactor. The regenerator off-gas is sent to a precipitator for removal of catalyst fines and then to a carbon monoxide boiler where CO is further oxidized and the heat of combustion recovered to produce process steam. Gas leaving the CO boiler is discharged to the atmosphere.[7]

Sulfur dioxide originates from the sulfur in the feedstocks and the auxillary fuel fired in the CO boiler. Typical regenerator emission levels range from 300 to 525 lb $SO_2/10^3$ bbl fresh feed,[7,8] and somewhat higher after the CO boiler depending on sulfur content and quantity of the auxillary fuel. The allowable SO_2 emission rate is determined by the process weight equation in §52.1881(b)(8) previously quoted.[3]

The Lima Refinery FCC unit processes 26,800 bbl/day of feedstock oil having a density of 7.53 lb/gal.[2] This results in a process weight of 176.6 ton/hr. The allowable SO_2 emission rate is 624.4 lb SO_2/hr. According to the Ohio Emissions Inventory Confirmation Report, the average SO_2 emission rate from the FCC/CO boiler is 298.6 lb SO_2/hr, well below the permissible level. Based on information obtained during the site visit,[2] GCA calculated an emission rate of 415 lb SO_2/hr, still in compliance with the proposed process regulation.

SECTION III

CONTROL TECHNOLOGY FOR DESULFURIZATION OF CLAUS PLANT TAIL GAS

Numerous Claus Plant tail gas desulfurization systems are in commercial operation in the United States today. A list of these processes is provided in Table 6. The technical capability of each process to meet the proposed Ohio SO_2 emission limitation for sulfur recovery plants is examined in the following sections. Three of the processes have demonstrated, in industrial use, their ability to meet compliance SO_2 levels. They are the Beavon process, the Wellman-Lord process and the SCOT process.

Table 6. DESULFURIZATION SYSTEMS FOR TREATING CLAUS PLANT TAIL GAS

System	Licensor	Licensed plants	
		U.S.	Worldwide
Beavon sulfur removal process	Union Oil Co. of California, Brea, California	26	31
IFP - 1500	Institut Francais du Pétrole	3	24
IFP - 150	Institut Francais du Pétrole	0	4
Shell Claus off-gas treating (SCOT) process	Shell Development Co., Houston, Texas	12	35
Sulfreen	Ralph M. Parsons Co., Pasadena, California	0	10
Wellman-Lard SO_2 recovery process	Davy Powergas, Inc., Lakeland, Florida	4	7

BEAVON SULFUR REMOVAL PROCESS[9,10,11]

The Beavon Sulfur Removal Process was developed jointly by the Ralph M. Parsons Company* and the Union Oil Company of California+ and is licensed by the Union Oil Company. A schematic diagram of the process is shown in Figure 2. The Claus Plant tail gas is heated to reaction temperature by mixing with the hot combustion products formed in the reducing gas (CO, H_2) generator. The combined gas stream is then passed through a cobalt-molybdate catalyst bed where all sulfur compounds are converted to H_2S by hydrogenation and hydrolysis according to the following reactions:[9]

$$S + H_2 \rightarrow H_2S$$

$$SO_2 + 3H_2 \rightarrow H_2S + 2H_2O$$

$$CH_3SH + H_2 \rightarrow H_2S + CH_4$$

$$CS_2 + 2H_2O \rightleftarrows 2H_2S + CO_2$$

$$COS + H_2O \rightleftarrows H_2S + CO_2$$

$$CO + H_2O \rightleftarrows CO_2 + H_2$$

After exiting the catalytic reactor, the gas stream is cooled in a waste heat boiler and/or quench tower. After cooling, the gases are fed to a Stretford absorber. The H_2S is absorbed in an aqueous solution of sodium carbonate, sodium metavanadate, and anthraquinone 2,7-disulfonic acid (ADA). The H_2S is initially absorbed in the alkaline solution per the following reaction:

$$H_2S + Na_2CO_3 \rightarrow Na^+ + HS^- + NaHCO_3$$

---

*Ralph M. Parsons Co., Pasadena, California 91124

+Union Oil Company of California, Brea, California 92621

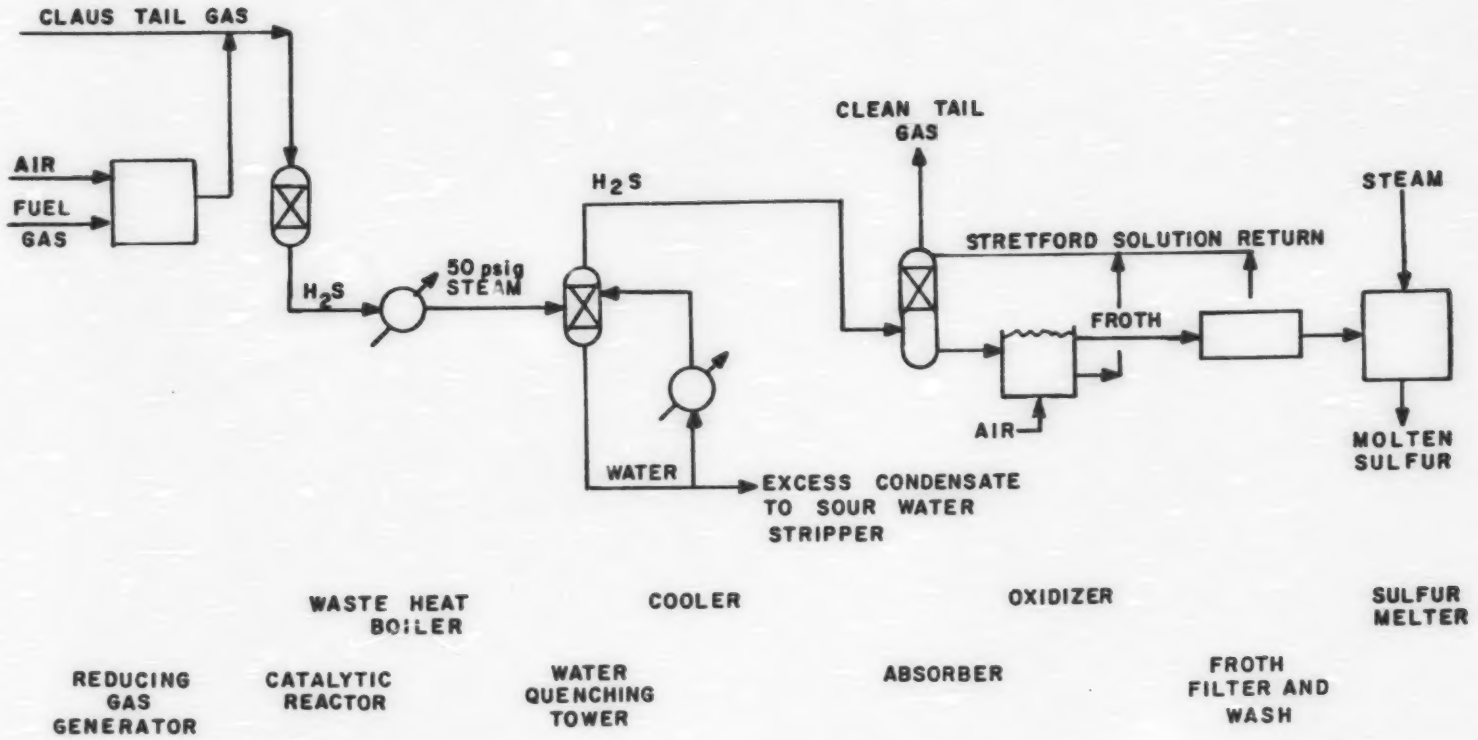

Figure 2. Flow diagram of the Beavon sulfur removal process

The HS^- ion then reacts with pentavalent vanadium to precipitate colloidal sulfur:

$$HS^- + 2V^{+5} \rightarrow S + 2V^{+4} + H^+$$

$$(NaHS + NaHCO_3 + 2NaVO_3 \rightarrow S + Na_2V_2O_5 + Na_2CO_3 + H_2O)$$

The reduced vanadium is subsequently oxidized back to the pentavalent state by blowing with air in the presence of the ADA which works as an oxidation catalyst:

$$Na_2V_2O_5 + 1/2\ O_2 \xrightarrow{ADA} 2NaVO_3$$

In the same operation, the finely divided sulfur appears as a froth which is skimmed off, filtered, and washed. The product sulfur is then separated from the washwater and melted. The Stretford solution is recirculated to the absorber from the oxidizer and from the sulfur filter.[9,10]

The treated tail gas exits the system from the absorber overhead. It is odorless, contains only a trace of H_2S and small amounts of COS and CS_2 which are not converted in the catalytic reactor. The concentration of all sulfur compounds combined is less than 100 ppm. Incineration of the off gas is not required.[11]

Results of emission tests performed by Environmental Science and Engineering, Inc. on the Beavon unit at Union Oil Co. of California's Los Angeles Refinery show that sulfur emissions are orders of magnitude below the allowable level.[12] These tests were conducted for the USEPA Office of Air Quality Planning and Standards. The results are presented in Table 7. Sulfur emissions (SO_2 equivalent) for the three days of testing averaged 0.00013 lb SO_2/lb sulfur processed. Note that the mean sulfur feed to the Claus Plant during the tests was

Table 7. SUMMARY OF EMISSION TEST DATA - BEAVON SULFUR RECOVERY UNIT[12]

Test date	Flow rate, scfm	Pollutant concentration,[a] ppmv, dry					Sulfur mass emission rate (SO_2 equivalent), lb/hr	Sulfur feed to Claus Plant, lb/hr	Emission level, lb SO_2/lb S
		SO_2	COS	H_2S	CS_2	TS[b]			
5 March 1974	2,313	1.5	17	<0.1	<0.15	18.9	0.44	3,145	1.41×10^{-4}
6 March 1974	2,529	0.7	17	<0.1	*	17.8	0.46	3,341	1.37×10^{-4}
7 March 1974	2,430	0.76	15	<0.1	*	15.86	0.39	3,127	1.25×10^{-4}
Average	2,424	1.0	16.3	<0.1	*	17.4	0.43	3,204	1.34×10^{-4}

[a] Determined using gas chromatograph

[b] Total sulfur as SO_2

* Not detectable

34.3 lt/day, very close to the sulfur feed rate to the Lima Refinery Claus Plant.

One inherent problem associated with the Stretford Process for recovering sulfur from H_2S is that some of the hydrogen sulfide undergoes side reactions and is converted to sodium sulfate and sodium thiosulfate.[9] Some sodium carbonate is also produced from the absorbed CO_2. The build-up of these by-products in the circulating Stretford solution necessitates continuous purging or periodic replacement of a portion of the solution. The purge stream contains no toxic materials, and as such presents no serious waste disposal problem. However, the purge does increase chemical consumption, which is reflected in the daily operating cost.[9]

Currently, 31 commercial Beavon units are in operation, under construction, or in design worldwide with 26 of the units in the United States. At least 16 of the units are operating, and reliability has been entirely satisfactory.[13]

SHELL CLAUS OFF-GAS TREATING (SCOT) PROCESS[14,15,16]

The SCOT process was developed by Shell Internationale Research Mij. B.V., The Netherlands, and is licensed in the United States by Shell Development Co.* The process is illustrated in the flow diagram, Figure 3.[14] The sulfur compounds in the Claus Plant tail gas are converted to hydrogen sulfide as in the Beavon process; i.e., the Claus off-gas is heated along with the addition of an H_2^- and CO-rich reducing gas and passed through a reactor containing a cobalt-molybdate catalyst. The gas is cooled utilizing a waste heat boiler for steam generation and a water quenching tower. Excess condensate from the quench is routed to an existing sour water stripper.[15]

*Shell Development Co., Houston, Texas.

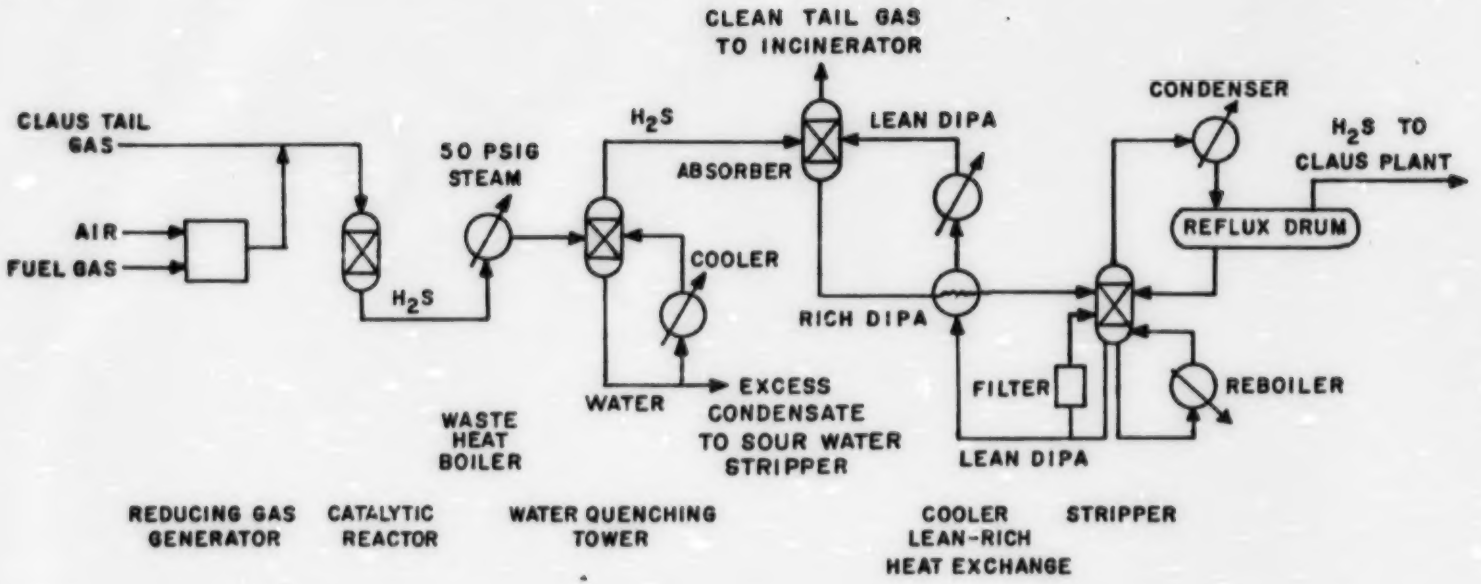

Figure 3. Flow diagram of SCOT process

Gas from the quench tower is counter-currently contacted in an absorption column with a diisopropanolamine (DIPA) solution which absorbs all but trace amounts of the H_2S plus about 30 percent of the CO_2. The sulfide-rich DIPA is sent to a conventional stripper where the H_2S is removed from the solution. The overhead gas, mostly H_2S, is recycled to the Claus recovery plant inlet. The lean amine solution is returned to the absorber. The absorber outlet gas, containing some unabsorbed H_2S and residual organic sulfides, is sent to the original Claus tail-gas incinerator before discharge to the atmosphere. Typical concentrations of SO_2 are in the 200 to 500 ppm range.[14]

Environmental Science and Engineering, Inc. performed emission tests on the SCOT unit operating at Champlin Petroleum Company's refinery at Wilmington, California, in March 1974.[16] The results of these tests are presented in Table 8. Total sulfur emissions (SO_2 equivalent) for the two days of testing averaged 0.0066 lb SO_2/sulfur processed, well below the allowable level even though upset conditions were encountered during the tests.

Thirty-five SCOT plants are under contract in the U.S., Canada, Europe, and Japan. At least 12 of these units are operating, 11 in the U.S.[14,15]

WELLMAN-LORD SO_2 RECOVERY PROCESS[17,18,19,20,21]

The Wellman-Lord SO_2 Recovery Process illustrated in Figure 4,[17] is available from Davy-Powergas, Inc.* Sulfur fed to the process must be in the form of sulfur dioxide. Therefore, Claus Plant tail gas must be incinerated. The burned gas stream is then cooled in a waste heat boiler and direct contact quench tower before entering an absorption column. In the absorber, the gas stream is scrubbed with a lean sodium

*Davy-Powergas, Inc., P. O. Box 2436, Lakeland, Florida 33803

Table 8. SUMMARY OF EMISSION TEST DATA - SCOT UNIT[16]

Test date	Flow rate, scfm	Pollutant concentration,[a] ppmv, dry					Sulfur mass emission rate (SO_2 equivalent), lb/hr	Sulfur feed to Claus Plant, lb/hr	Emission level, lb SO_2/lb S
		SO_2	COS	H_2S	CS_2	TS[b]			
14 March 1974	2,305	220	18	*	< 1.7	240	5.62	1,195	0.0047
15 March 1974	4,080	118	56	58	<11.5	255	9.91	1,232	0.0080
Average	3,192	169	37	29	< 6.6	248	8.04	1,213	0.0066

[a] Determined using gas chromatograph

[b] Total sulfur as SO_2

* Indeterminate

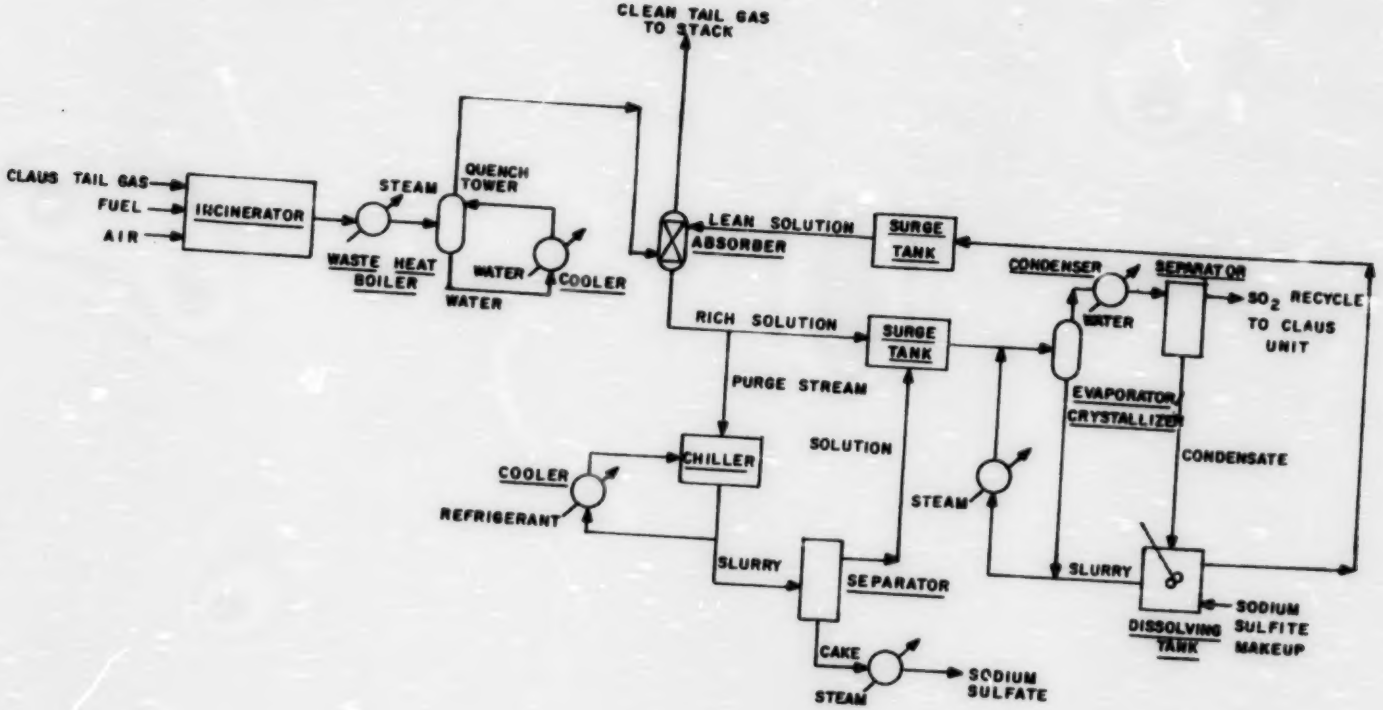

Figure 4. Wellman-Lord SO_2 recovery process flow diagram

sulfite (Na_2SO_3) solution to form sodium bisulfite ($NaHSO_4$) and sodium pyrosulfite ($Na_2S_2O_5$) per the following reactions:[17,18]

$$SO_2 + Na_2SO_3 + H_2O \rightarrow 2NaHSO_3$$

$$2NaHSO_3 \rightarrow Na_2S_2O_5 + H_2O$$

The clean gas exits the absorber and is discharged to the atmosphere, typically at SO_2 concentrations of less than 100 ppm.[19]

The rich bisulfite solution is fed to an evaporator/crystallizer regeneration system. Heat supplied to the evaporator causes the decomposition of the bisulfite and pyrosulfite:

$$2NaHSO_3 \rightarrow Na_2SO_3 + SO_2 + H_2O$$

$$Na_2S_2O_5 \rightarrow SO_2 + Na_2SO_3$$

The SO_2 and water vapor pass overhead from the evaporator to a primary condenser, where 80 to 90 percent of the water is removed. The condensate is stripped with steam to remove dissolved SO_2. Stripper overhead and condenser overhead vapors are mixed and sent to a secondary condenser. Condensate is returned to the stripper and the vapor stream is recycled to the Claus Plant. The recycle is 90 to 95 percent SO_2.[19]

The main problem facing the Wellman-Lord process is the formation of nonregenerable sulfate and thiosulfate by the following side reactions:[18,21]

$$Na_2SO_3 + 1/2\ O_2 \rightarrow Na_2SO_4$$

$$SO_2 + 3Na_2SO_3 \rightarrow 2Na_2SO_4 + Na_2S_2O_3$$

$$Na_2SO_3 + S \rightarrow Na_2S_2O_3$$

$$2Na_2SO_3 + SO_3 \rightarrow Na_2SO_4 + Na_2S_2O_5$$

This creates the need to purge a portion of the solution from the system, necessitating chemical replacement and posing a waste disposal problem. Chemical requirements have been minimized by a system which selectively removes the sulfate by crystallization. The concentrated purge stream can then be dried for sale or disposal. Davy-Powergas is currently engaged in the development of processes to chemically reduce the sulfate back to sodium sulfite.[17]

There are now 17 Wellman-Lord installations in commercial service in the United States and Japan, desulfurizing tail gas from Claus plants, sulfuric acid plants, and boilers.[22] An additional 13 plants are in the design or construction phase. Five of the operating units treat Claus Plant tail gas. One such unit, located at Standard Oil of California's El Segundo Refinery, was selected by the USEPA Office of Air Quality Planning and Standards for an emission testing program. Results of the tests, conducted by Environmental Sciences and Engineering, Inc., are reported in Table 9.[21] Average emissions for the 3 days of testing were 1.7×10^{-4} lb SO_2/lb sulfur processed. Total recoverable sulfur to the Claus unit was 112.3 lt/day.

SULFREEN PROCESS[23,24,25]

The Sulfreen process was developed by Lurgi Apparate-Technik, Frankfurt, West Germany, in conjunction with France's Société Nationale des Pétroles d'Aquitaine.[23] The licensor in the United States is the Ralph M. Parsons Co. A flow diagram of the process is provided in Figure 5.[24]

The process is essentially an extension of the Claus process, except that the H_2S and SO_2 react at temperatures below the sulfur dew point.

Table 9. SUMMARY OF EMISSION TEST DATA - WELLMAN-LORD SO_2 RECOVERY UNIT[21]

Test date	Flow rate, scfm	Pollutant concentration,[a] ppmv, dry					Sulfur mass emission rate (SO_2 equivalent), lb/hr	Sulfur feed to Claus Plant, lb/hr	Emission level, lb SO_2/lb S
		SO_2	COS	H_2S	CS_2	TS[b]			
11 March 1974	6,960	38	3.2	<0.1	2.5	46.2	3.27	10,920	0.00030
12 March 1974	4,782	16	1.9	<0.1	3.4	24.7	1.20	8,400	0.00014
13 March 1974	7,370	10	0.91	<0.1	1.1	13.1	0.98	12,133	0.00008
Average	6,371	21.3	2.0	<0.1	2.3	28	1.81	10,484	0.00017

[a] Determined by gas chromatograph

[b] Total sulfur as SO_2

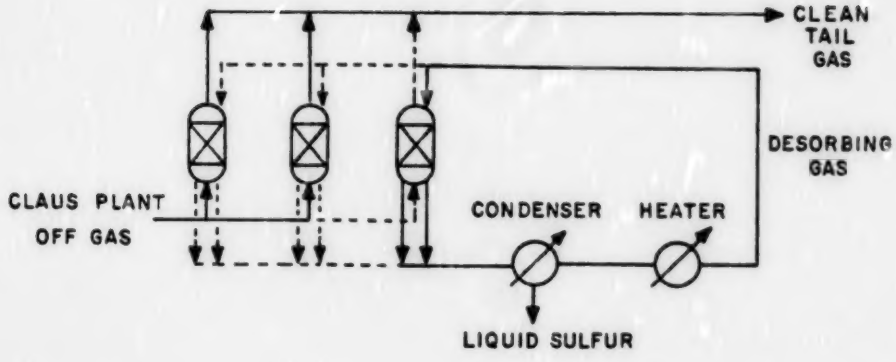

Figure 5. Sulfreen process flow diagram

The reaction is as follows:

$$2H_2S + SO_2 \rightarrow 3S + 2H_2O$$

As temperature is lowered, equilibrium is shifted to the right, and conversion becomes more complete. The reaction takes place in the presence of an activated alumina catalyst.

Sulfur is adsorbed on the catalyst, requiring periodic regeneration of the catalyst bed by desorbing the sulfur with hot gas. Hence, multiple reactors are necessary to provide continuous operation. At any given time, two-thirds of the reactors are in the absorption phase and the remaining one-third is in the regeneration phase. Desorption gas (usually nitrogen) containing sulfur vapor is sent to a condenser where the sulfur is removed and combined with the Claus produced sulfur.[23,24]

The overhead gas from the catalytic reactors is emitted to the atmosphere. As evident from the emission test results summarized in Table 10,[25] the Sulfreen process does not sufficiently lower the sulfur emissions to meet the proposed Ohio standards. Average sulfur emission (SO_2 equivalent) for the three days of testing was 0.038 lb SO_2/lb of sulfur processed.[25]

IFP-1500 PROCESS[26,27,28]

The IFP-1500 process was developed by the Institut Francais du Pétrole* specifically for the removal of sulfur compounds from Claus Plant tail gas. The technology involved is essentially an extension of the Claus reaction, but carried out in the liquid phase. As illustrated in Figure 6,[26] Claus Plant tail gas is injected into the bottom of a packed

*Institute Francais du Pétrole, New York, N.Y. 10022

Table 10. SUMMARY OF EMISSION TEST DATA - SULFREEN UNIT[25]

Test date	Flow rate, scfm	Pollutant concentration,[a] ppmv, dry					Sulfur mass emission rate (SO_2 equivalent), lb/hr	Sulfur feed to Claus Plant, lb/hr	Emission level, lb SO_2/lb S
		SO_2	COS	H_2S	CS_2	TS[b]			
24 Sept. 1974	148,260	3,800	1,100	3,520	800	10,020	15,092	327,787	0.046
25 Sept. 1974	143,680	3,750	750	3,870	650	9,670	14,114	365,493	0.039
27 Sept. 1974	144,375	1,700	750	3,590	800	7,640	11,205	374,267	0.030
Average	145,438	3,083	867	3,660	750	9,110	13,470	355,849	0.038

[a] Determined using gas chromatograph

[b] Total sulfur as SO_2

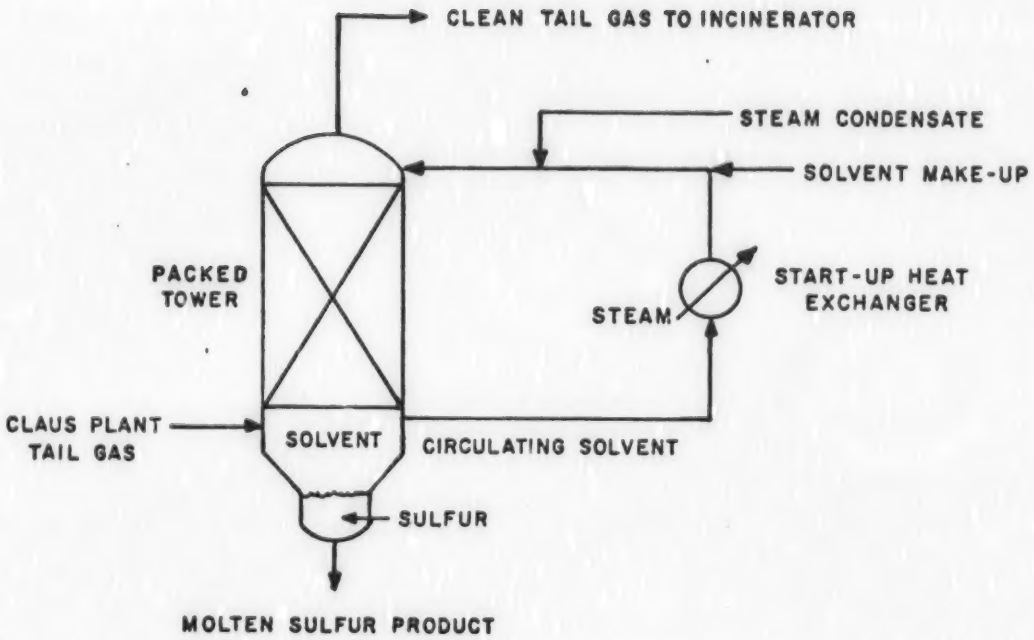

Figure 6. IFP-1500 Claus tail-gas treatment

tower and contacted counter-currently with a catalyst-containing solvent, polyethylene glycol. The catalyst is a carboxylic acid salt which forms a complex with H_2S and SO_2. Elemental sulfur is generated by the Claus reaction. Heat is removed by injecting and vaporizing steam condensate. Overhead gases are sent to the incinerator before release to the ambient air.[26,27]

As reported in Table 11,[28] emission test data gathered from an IFP-1500 unit on a Claus Plant operated by Stauffer Chemical Company in Delaware City, Delaware, show that the IFP-1500 system will not reduce sulfur emissions sufficiently to comply with the proposed Ohio limitation. The average emission level was 0.0111 lb SO_2/lb S processed.[28]

IFP-150 PROCESS[26,27]

The Institut Francais du Pétrole also licenses a second process for reducing Claus Plant emissions, the IFP-150. A flow diagram of the process is provided in Figure 7.[26]

The IFP-150 process incorporates the IFP-1500 process with an ammonia scrubbing section. The Claus tail gas is incinerated, then scrubbed with aqueous ammonia to reduce SO_2 concentration to 150 to 250 ppm range. The brine, containing mostly ammonium sulfites, is heated and sent to an evaporator where the sulfites decompose to ammonia and water. The overhead gas stream is mixed with an acid gas stream and sent to the catalytic reactor. The evaporator bottoms containing thermostable sulfates and thiosulfates, are reduced in a proprietary process to ammonia, sulfur dioxide, and water which are then mixed with the overhead stream.[26,27]

In the reactor tower, H_2S and SO_2 dissolve in polyethylene glycol and react to form elemental sulfur and water in a process similar to the IFP 1500 system. The overhead gas is composed of ammonia and water, which are condensed and returned to the ammonia scrubber.

Table 11. SUMMARY OF EMISSION TEST DATA - IFP-1500 PROCESS[28]

Test date	Flow rate, scfm	Pollutant concentration,[a] ppmv, dry					Sulfur mass emission rate (SO_2 equivalent), lb/hr	Sulfur feed to Claus Plant, lb/hr	Emission level, lb SO_2/lb S
		SO_2	COS	H_2S	CS_2	TS[b]			
10 June 1974	14,867	82	132	1,190	460	2,324	351.0	*	*
11 June 1974	15,220	72	77	1,410	180	1,919	296.7	*	*
12 June 1974	14,620	80	133	1,950	300	2,763	410.4	*	*
Average	14,902	78	114	1,517	313	2,335	352.7	31,710	0.0111

[a] Determined using gas chromatograph

[b] Total sulfur as SO_2

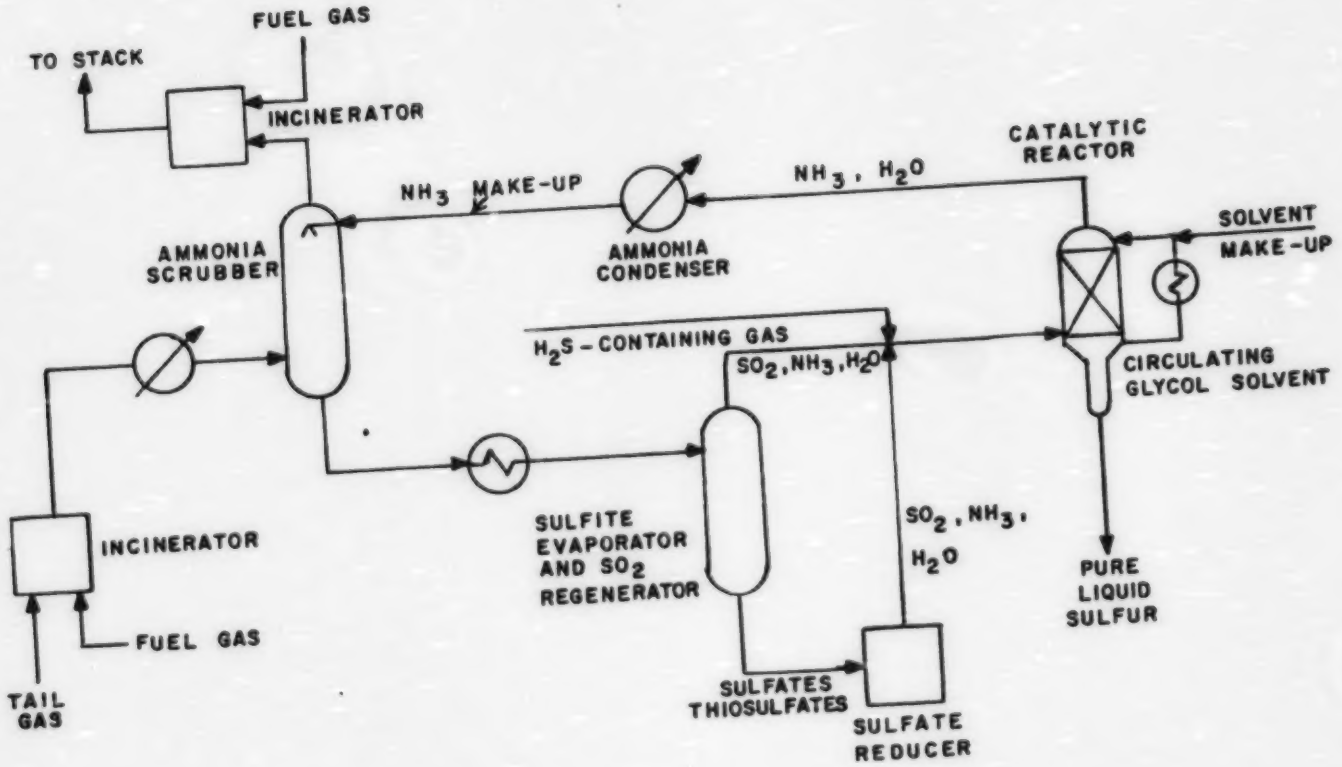

Figure 7. Flow diagram of IFP-150 process[26]

Four IFP-150 units are under construction or already operating in Japan and France. However, no units have yet been built in the United States. A Claus unit equipped with IFP-150 tail gas treatment should be able to meet the proposed Ohio SO_2 emission limitation for sulfur recovery plants.

In summary, six commercially available Claus tail gas desulfurization processes were reviewed to ascertain the technical capability of complying with the proposed Ohio SO_2 emission regulation. Three of the processes have clearly demonstrated this capability - the Beavon process, the Wellman-Lord process, and the SCOT process. The IFP-150 process should also be capable of meeting the proposed 0.01 lb SO_2/lb S regulation. In Section IV, the estimated costs of installing and operating a Beavon unit or a SCOT unit at the SOHIO Lima Refinery are presented.

SECTION IV

COST OF CONTROL SYSTEMS

GCA has prepared detailed cost estimates for Claus plant SO_2 emission control at SOHIO's Lima Refinery. Estimates were made for a Beavon system and for a SCOT system. The costs apply to a system designed according to the specifications in Section II. Battery-limits plant cost and utility, chemical, and labor requirements were supplied by the engineering-construction firms that design and install the units.

A current estimate of the capital and operating costs for an entire fuel gas desulfurization and sulfur recovery system comparable to that in use at the Lima Refinery was also prepared. The estimate is based on cost data received from SOHIO[2] updated to June 1976 prices using the Marshall and Swift equipment cost index.[29]

The estimated capital requirement for the desulfurization and sulfur recovery system without tail gas cleanup is $8.6 million with an annual cost of $3.3 million. The cost of retrofit Claus tail gas desulfurization systems are $2.0 million and $2.1 million for a Beavon system and SCOT system, respectively. The associated annual costs are $541 thousand and $652 thousand.

COST OF CLAUS PLANT SO_2 EMISSION CONTROL

To obtain accurate equipment and installation costs, GCA asked two engineering-construction firms for cost quotations on a battery-limits desulfurization facility for treating the tail gas from the Lima Refinery

Claus unit. The two firms were Ford, Bacon, and Davis, Inc. of Dallas, Texas, a vendor of the SCOT process; and The Ralph M. Parsons Co., of Pasadena, California, who designs and builds Beavon units. Each company supplied battery-limit plant costs and utility, chemical, and manpower requirements.[13,30]

The average daily usage of utilities, materials, and labor is reported in Table 12. Also presented are the utility, material, and labor costs derived from these operating requirements. The SCOT process uses steam for stripping purposes while the Beavon process does not require steam. Both systems receive some credit for steam produced in waste heat boilers. Chemicals used in the Beavon process are lost in the purge stream, the oxidizer air stream, and a small amount in the product sulfur. Di-isopropanolamine (DIPA) used as the absorbent in the SCOT process is lost in the sludge blowdown and carryover from the stripper. In the absence of hard data on maintenance materials, the cost was assumed to be 1.5 percent of the fixed capital investment per year.

The equipment vendors reported that one operator per shift is required to handle normal operations. In addition, a laboratory technician is required 2 hours/day to run periodic analyses. Maintenance labor is assumed to be equal to the cost of maintenance materials.

The capital requirements, annualized costs, and the components used to evaluate these costs are presented in Table 13. The annualized costs were derived according to an accounting method based on the Utility Financing Method and modified by the Panhandle Eastern Pipeline Company.[31] These costs are current as of June 1976. The battery-limit plant costs were furnished by the engineering-construction firms.[13,30] The total capital investment was amortized over 20 years at a discounted cash flow rate of 8 percent, and the total annual operating costs were derived primarily from Table 12.

The estimated capital requirement for the Beavon system was $1,987,000 with an attendant average annual cost of $541,000. A SCOT system for the

Table 12. UTILITY, MATERIAL, AND LABOR REQUIREMENTS AND COSTS FOR BEAVON AND SCOT CLAUS TAIL GAS TREATMENT SYSTEMS AT SOHIO'S LIMA REFINERY[13,30]

	Average daily usage			Average daily cost	
	Beavon process (Ralph M. Parsons Co.)	SCOT process (Ford, Bacon, and Davis)	Unit cost	Beavon process	SCOT process
Utilities					
Power	3,600 kWh	1,200 kWh	$0.02/kWh[a]	$ 72.0	$ 24.0
Steam	32,900 lbs	180,000 lbs	$2/1000 lbs[b]	65.8	360.0
Fuel gas	57.6 MM Btu	43.2 MM Btu	$1/MM Btu	57.6	43.2
Steam produced	38,400 lbs	38,400 lbs	$2/1000 lbs	$195.4	$427.2
Total				-76.8	-76.8
				$118.6	$350.4
Materials					
Anthraquinone disulfonic acid (ADA)	6.6 lbs	-	$5.3/lb[c]	$ 35.0	-
Vanadium	9.2 lbs	-	$5/lb[c]	46.0	-
Sodium carbonate	368.0 lbs	-	$0.1/lb[c]	36.8	-
Diisopropanolamine (DIPA)	-	250 lbs	$0.44/lb[d]	-	$110.0
Maintenance	-	-	1.5% of fixed capital investment per year[e]	78.9	84.6
Total				$196.7	$194.6
Labor					
Operator	24 hr	24 hr	$8/hr	$192.0	$192.0
Chemist	2 hr	2 hr	$8/hr	16.0	16.0
Maintenance	-	-	1.5% of fixed capital investment per year[e]	78.9	84.6
Total				$286.9	$292.6
TOTAL				$602.2	$837.6

[a] Ohio Edison

[b] Based on electricity cost and 10 lb steam/kWh

[c] Personal communication with Mr. W. E. Carbone of Wilputte Corporation, 152 Floral Avenue, Murray Hill, N. J. 07974, May, 1976

[d] Chemical Marketing Reporter, June 21, 1976

[e] GCA assumption, similar to estimate used by Choi, et al.,[31] for industrial pollution control processes

Table 13. COSTS FOR BEAVON AND SCOT CLAUS TAIL GAS TREATMENT AT SOHIO'S LIMA REFINERY, 10^3, JUNE 1976

	Beavon	SCOT
Capital requirement		
Battery limits plant installed (BLP)[a]	$1,600.0	$1,715.0
Site preparation and utilities,[b] 20% of BLP	320.0	343.0
Fixed capital investment (FCI)	1,920.0	2,058.0
Working capital,[c] 20% of gross operating cost	66.9	85.1
Total capital investment	$1,986.9	$2,143.1
Annual operating cost		
Labor[d]	$ 104.7	$ 106.8
Administrative and general overhead,[c] 60% of labor	62.8	64.1
Utilities[d]	43.3	127.9
Materials[d]	71.8	71.0
Local taxes and insurance,[c] 2.7% of FCI	51.8	55.6
Gross operating cost	334.4	425.4
Sulfur by-product credit, $45/long ton	-31.4[e]	-30.6[f]
Net operating cost	$ 303.0	$ 394.8
Annualized cost		
Annualized capital cost[g]	$ 202.4	$ 218.3
Federal income tax[c,h]	35.6	38.6
Net operating cost	303.0	394.8
Average annual cost	$ 541.0	$ 651.7
Average annual cost, $/lb S removed	$ 0.346	0.429

[a] Vendor estimates.[13,30]

[b] Site preparation, cost to bring utilities to the battery limits of plant. Based on review of similar costs in the literature.

[c] Based on Utility Financing Method modified by the Panhandle Eastern Pipeline Company and described in Reference 31.

[d] See Table 12.

[e] Based on $45/LT currently received by SOHIO from sulfuric acid manufacturer and 50 ppm SO_2 out stack.

[f] Based on 300 ppm SO_2 out stack

[g] Capital costs are amortized at a discount cash flow rate of 8 percent over 20 years. This method yields an average capital cost, including depreciation, of 10.185 percent of total capital investment.

[h] Average federal income tax = 1.731 percent of sum of total capital investment and working capital.

Lima Refinery would require a total capital outlay of $2,143,000 with an annual cost of $652,000. The average annual control cost, $1b/sulfur removed, is $0.346 for the Beavon system and $0.429 for the SCOT system.

COST OF FUEL GAS DESULFURIZATION AND SULFUR RECOVERY

The capital requirement and annual operating costs (June 1976) to build and operate a desulfurization and sulfur recovery system comparable to that in use at the Lima Refinery are presented in Table 14. An estimated capital outlay of $8.6 million is required with an annualized cost of $3.3 million. These estimates are based on cost data received from SOHIO pertaining to the purchase, installation, and operation of the MEA and DEA fuel gas desulfurization units, Claus plant, and foul condensate stripper.[2] The average control cost, $/1b, sulfur removed, is $0.121.

Addition of a Beavon or SCOT system for treating the Claus plant tail gas will increase the total capital requirement to $10.6 million and $10.7 million, respectively. Annual operating costs become $3.9 million to $4.0 million, with the average control cost equal to $0.133/1b sulfur removed with the Beavon system and $0.137 with the SCOT system.

Table 14. ESTIMATED CAPITAL REQUIREMENT AND ANNUALIZED COSTS FOR A FUEL GAS DESULFURIZATION AND SULFUR RECOVERY SYSTEM COMPARABLE TO THE LIMA REFINERY SYSTEM, 10^3, JUNE 1976

Capital requirement	
Fixed capital investment[a]	$8,000
Working capital, 20% of gross operating cost[b]	568
Total capital investment	$8,568
Annual operating cost	
Labor[c]	$ 262
Administrative and general overhead, 60% of labor	157
Utilities[c]	1,953
Materials[c]	250
Local taxes and insurance, 2.7% of FCI[b]	216
Gross operating cost	$2,838
Sulfur by-product credit,[d] $45/long ton	552
Net operating cost	$2,286
Annualized cost	
Annualized capital cost[e]	$ 873
Federal income tax[b,f]	158
Net operating cost	2,286
Average annual cost	$3,317
Average control cost, $/lb S removed	0.121

[a]Based on capital cost supplied by SOHIO and updated according to the Marshall & Swift index.[29]

[b]Based on Utility Financing Method modified by the Panhandle Eastern Pipeline Company and described in Reference 31.

[c]Based on costs supplied by SOHIO.

[d]Based on design sulfur recovery rate of 33.6 long ton/day.

[e]Capital costs are amortized at a discount cash flow rate of 8 percent over 20 years. This method yields an average capital cost, including depreciation, of 10.185 percent of total capital investment.

[f]Average federal income tax = 1.731 percent of sum of total capital investment and working capital.

SECTION V

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